

Abstract

We have used polarized neutron reflectometry to individually examine the magnetization

reversals of ferromagnetic Ga_{1-x}Mn_xAs layers separated by a non-magnetic GaAs spacer

layer of varying thickness. For each of the samples studied, the top Ga_{1-x}Mn_xAs layer is

adjacent to a Be-doped Al_{0.25}Ga_{0.75}As capping layer on one side and the GaAs spacer on

the other, while the bottom $Ga_{1-x}Mn_xAs$ layer is surrounded by GaAs on either side. For

samples with spacer thicknesses of 12 and 6 nm, antiparallel alignment of the two Ga₁₋

_xMn_xAs layer magnetizations was observed at multiple fields, implying that hole doping

from the capping layer strongly affects the coercivity of the top $Ga_{1-x}Mn_xAs$ layer, but has

a weaker effect on the coercivity of the bottom $Ga_{1-x}Mn_xAs$ layer. However, for a spacer

thickness of 3 nm, both top and bottom $Ga_{1-x}Mn_xAs$ layers appear to be equally

influenced by the capping layer, as virtually identical coercivities were observed. This

behavior is evidence of coupling between the Ga_{1-x}Mn_xAs layers across the 3 nm GaAs

spacer.

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For development of many potential spintronic devices, it is desirable to have semiconductor materials with true long range ferromagnetic (FM) order. Ga_{1-x}Mn_xAs has been shown to be just such a material, with FM originating from a hole mediated exchange. 1,2 Interlayer coupling in magnetic multilayer structures is a phenomenon exploited with great utility in numerous device applications,³ and it is therefore important for spintronics researchers to understand such coupling in Ga_{1-x}Mn_xAs-based multilayer devices. SQUID magnetometry, 4,5,6,7 magneto-transport measurements, 4,5,7 and qualitative analysis of neutron diffraction superlattice peaks^{8,9} have been used to indirectly infer evidence of interlayer coupling between separated Ga_{1-x}Mn_xAs layers. However, the magnetic and structural properties of *individual* layers in a multilayer structure can be directly obtained through a quantitative analysis of the structure's polarized neutron reflectivity (PNR). 10,11,12,13 We have previously reported our use of this technique to precisely determine the structural profiles and temperature (T) dependent magnetizations of a series of samples in which two Ga_{1-x}Mn_xAs layers made to have different (in the absence of any interlayer coupling) Curie temperatures (T_C) and coercive fields (H_C) are separated by a non-magnetic GaAs spacer layer of varying thickness. ¹⁴ Here, we report on PNR measurements of the same samples performed as function of applied magnetic field (H). We observe that for 12 and 6 nm spacers, the two Ga_{1-x}Mn_xAs layers have very different coercivities, but that for a 3 nm spacer the coercivities of the two layers are virtually identical.

Three 1 cm x 2 cm rectangular samples were prepared by molecular beam epitaxy on GaAs substrates¹⁵ with the following layer structure (starting at the substrate interface):

- 16 nm bottom Ga_{0.95}Mn_{0.05}As layer,
- 12, 6, or 3 nm GaAs spacer,
- 8 nm top $Ga_{0.95}Mn_{0.05}As$ layer,
- 25 nm Al_{0.25}Ga_{0.75}As cap doped with Be at a concentration of 3 x 10²⁰ cm⁻³.

Through modulation doping, the Be-doped capping layer is a source of extra holes for the adjacent top $Ga_{1-x}Mn_xAs$ layer. The addition of holes affects the hole-mediated FM exchange in $Ga_{1-x}Mn_xAs$, and has been shown to reduce the coercivity. Since the bottom $Ga_{1-x}Mn_xAs$ layer has no adjacent source of extra holes, it will exhibit a M(H) curve very different from that of the top $Ga_{1-x}Mn_xAs$ layer – unless the two layers are coupled across the spacer.

PNR measurements were conducted using Asterix¹² at the Lujan Neutron Scattering Center of Los Alamos National Laboratory, and the NG-1 Polarized Beam Reflectometer¹⁹ at the NIST Center for Neutron Research. For these measurements, a neutron beam was polarized alternately spin-up (+) and spin-down (-) relative to H applied along the in-plane magnetic hard [110] sample direction, and was shined on the sample. The non spin-flip specular reflectivities R^{++} and R^{--} were measured as a function of wavevector transfer Q^{20} . A sample's depth-dependent nuclear scattering length density $\rho(z)$, and the component of the depth-dependent magnetization parallel to H, M(z), can be deduced by model fitting²¹ of $R^{++}(Q)$ and $R^{--}(Q)$. ^{10,11,12,13} In this way, we individually determined the component of the magnetization parallel to H for the Ga₁. ${}_xMn_xAs$ layer next to the Be doped AlGaAs cap (M_{Top}) and that for the Ga_{1-x}Mn_xAs layer next to the GaAs substrate (M_{Bot}) for each of the samples studied. Structural parameters

in the models such as layer thickness, interlayer roughness, and Mn concentration *x* were determined from the high resolution PNR measurements discussed in Ref. 14. These measurements confirmed the presence of non-magnetic spacer layers in each of the samples, and revealed that the samples are practically identical in structure and composition except for the thickness of the spacer layer.

H-dependent PNR measurements for each of the samples were conducted after cooling the sample to 6 K in H = +100 mT, lowering H = -100 mT, and then raising H = 0. Since the differences between $R(Q)^{++}$ and $R(Q)^{--}$ are due to M(z) it is intuitive to express the PNR data as spin asymmetry:

$$A(Q) = \frac{R^{++}(Q) - R^{--}(Q)}{R^{++}(Q) + R^{--}(Q)}.$$

Model calculations show that A(Q) corresponding to both M_{Top} and M_{Bot} being negative with respect to H should only differ from the A(Q) corresponding to both M_{Top} and M_{Bot} being positive with respect to H by a factor of -1. Thus, if both top and bottom Ga_1 . ${}_xMn_xAs$ layers reverse in the same way, as H is increased the amplitude of the A(Q) peaks will shrink to zero and then reverse in sign, but no change in the frequency of the oscillations will be observed. However, if M_{Top} and M_{Bot} reverse independently of one another, an antiparallel alignment of M_{Top} and M_{Bot} is observable as a A(Q) with non-zero amplitude and a frequency distinct from the frequency corresponding to parallel alignment of M_{Top} and M_{Bot} . Figure 1 shows example A(Q) data and fits for the samples at selected fields as A(Q) with an increased from -100 mT at A(Q) data at 1.4 mT (a) differs from the data at 12.5 mT (c) approximately by a factor of (-1). Model fitting reveals that these two data sets

correspond to parallel alignment of both M_{Top} and M_{Bot} , positively and negatively aligned with respect to H, respectively. However, at 9 mT (panel b) the A(Q) frequency is clearly different from that in panels a or b. Further, the lowest QA(Q) peak has almost zero amplitude, while large amplitudes are observed for higher Q peaks. Since smallest-Q corresponds to the largest length scales, this immediately suggests that the average M of the *entire* sample has approached zero, but that there are local regions of nonzero M. Indeed, quantitative analysis bears this out, as fitting shows that this data corresponds to anti-parallel alignment of M_{Top} and M_{Bot} . PNR data from the 6 nm spacer sample (panels d-f) reveals similar behavior, with evidence of anti-parallel alignment of M_{Top} and M_{Bot} . However, the 3 nm spacer sample (g-i) is different. Data corresponding to parallel alignment of both M_{Top} and M_{Bot} , positive and negative with respect to H, is observed at 1.8 mT (g) and 6.1 mT (i), respectively. But, no evidence of an anti-parallel alignment of M_{Top} and M_{Bot} was found at intermediate fields. Instead, A(Q) with nearly zero amplitude was observed at H = 3.8 mT, indicating near zero magnetization parallel to H for both $Ga_{1-x}Mn_xAs$ layers.

Fitting results are summarized in Figure 2, which shows the field dependencies of the individual layer magnetizations for each of the samples. For both the 12 (a) and 6 nm (b) spacer samples, the top $Ga_{1-x}Mn_xAs$ layer is observed to have a significantly smaller H_C (~ 4 mT) than the bottom $Ga_{1-x}Mn_xAs$ layer (~ 10 mT), as anti-parallel alignment of the layers is observed at multiple field values. This implies that for these two samples, the extra holes supplied by the Be doped AlGaAs cap strongly affect the magnetic exchange of the top $Ga_{1-x}Mn_xAs$ layer, but have a weaker influence over the exchange in the bottom

 $Ga_{1-x}Mn_xAs$ layer. For the 3 nm spacer sample the situation is different, as *both* the top and bottom $Ga_{1-x}Mn_xAs$ layers have $H_C \approx 4$ mT. Since this value of H_C is the same as that observed for the top layers of the samples with thicker spacers, this suggests that the influence of the capping layer extends across the 3 nm nonmagnetic GaAs spacer layer, and affects the magnetic exchange of both top and bottom $Ga_{1-x}Mn_xAs$ layers. Therefore, we conclude that the top and bottom $Ga_{1-x}Mn_xAs$ layers are strongly coupled when the spacer between them is 3 nm, and that the coupling weakens as the spacer thickness is increased.²⁴

These results constitute evidence that Ga_{1-x}Mn_xAs can strongly interact across a non-magnetic spacer layer, a property that may prove important for device applications. While we cannot determine the exact nature of the coupling from our results, we speculate that it could be due to RKKY-like magnetic exchange coupling²⁵, and/or an electronic coupling where carrier wavefunctions in the top Ga_{1-x}Mn_xAs layer overlap with those in the bottom Ga_{1-x}Mn_xAs layer, resulting in similar hole concentrations for the two layers.^{26,27} This work was supported by NSF DMR-0603752. The authors thank Mike Fitzsimmons of Los Alamos National Laboratory for assistance with Asterix measurements.

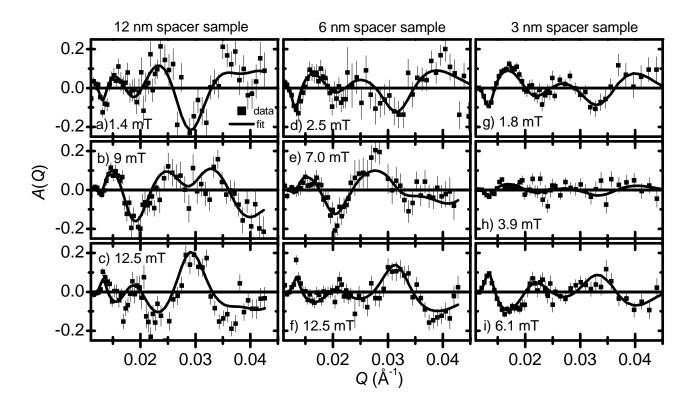


Figure 1: PNR data (symbols) and fits (lines) shown as spin asymmetry, A(Q) as selected fields as H is increased from -100 mT. The 12 nm spacer sample (a-c), and the 6 nm spacer sample data (d-f) show evidence of anti-parallel alignment of M_{Top} and M_{Bot} . The 3 nm spacer sample data (g-i) shows evidence that M_{Top} and M_{Bot} both approach zero at approximately the same field value.

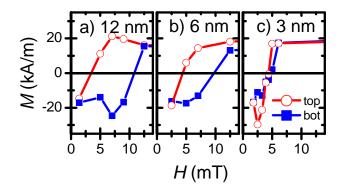


Figure 2: The M component parallel to H as a function of H for the top and bottom Ga_1 . ${}_xMn_xAs$ layers in a) the 12, b) the 6, and c) the 3 nm spacer samples, as determined from PNR. Solid lines are guides to the eye.

¹ B. Beschoten, P. A. Crowell, I. Malajovich, D. D. Awschalom, F. Matsukura, A. Shen, and H. Ohno, *Phys. Rev. Lett.* **83**, 3073 (1999).

² J. Blinowski and P. Kacman, *Phys. Rev. B* **67**, 121204(R) (2003).

³ G.A. Prinz, *Science* **282**, 1660 (1998).

⁴ N. Akiba, et al, **73**, 2122 (1998).

⁵ D. Chiba, et al, Appl. Phys. Lett. 77, 1873 (2000).

⁶ R. Mathieu, et al, Appl. Phys. Lett. **81**, 3013 (2002).

⁷ S.J. Chung, et al, J. Appl. Phys. **95**, 7402 (2004).

⁸ H. Kępa, et al, Phys. Rev B **64**, 121302(R) (2001).

⁹ W. Szuszkiewicz, et al, Acta Physica Polonica A, **100**, 335 (2001).

¹⁰ G.P. Felcher, *Phys. Rev. B* **24**, 1595 (1981).

¹¹ C.F. Majkrzak, *Physica B* **173**, 75 (1991).

¹² M.R. Fitzsimmons and C.F. Majkrzak, in *Modern Techniques for Characterizing Magnetic Materials*, edited by Z. Zhu (Kluwer, New York, 2005).

¹³ C.F. Majkrzak, K.V. O'Donovan, and N.F. Berk, in Neutron Scattering from Magnetic Materials, edited by T. Chatterji (Elsevier, New York, 2006).

¹⁴ B.J. Kirby, J.A. Borchers, X. Liu, Z. Ge, Y.-J. Cho, J.K. Furdyna, submitted to Physical Review B. Preprint available at http://www.arxiv.org: arXiv:0708.2289v2 [cond-mat.mtrl-sci].

¹⁵ T. Wojtowicz, et al, Appl. Phys. Lett. **83**, 4220 (2003).

¹⁶ X. Liu, et al, Phys. Rev. B 71, 35307 (2005).

¹⁷ B. Beschoten, et al, Phys. Rev. Lett. **83**, 3073 (1999).

¹⁸ S.J. Potashnik, K.C. Ku, R.F. Wang, M.B. Stone, N. Samarth, P. Schiffer, and S.H. Chun, *J. Appl. Phys.* 93, 6784 (2003).

¹⁹ http://www.ncnr.nist.gov/instruments/ng1refl/

²⁰ Spin-flip scattering, which is sensitive to the component of M(z) perpendicular to H, was not measured. Calculations show that due to the small magnitude of M, any spin-flip scattering from our samples would be weak enough to make such measurements impractical.

²¹ P.A. Kienzle, K.V. O'Donovan, J.F. Ankner, N.F. Berk, C.F. Majkrzak; http://www.ncnr.nist.gov/reflpak. 2000-2006.

²² Or one layer with zero M and the other layer with nonzero M.

 $^{^{23}}$ R^{++} and R^{--} were fit simultaneously fit, and plotted as A.

²⁴ Reference 14 shows evidence that, while weaker than for the 3 nm spacer sample, there is also interlayer coupling for the 6 nm spacer sample.

²⁵ P. Sankowski, and P. Kacman, *Phys. Rev. B* **52**, 201303(R) (2005).

²⁶ Z. Ge, Y-Y. Zhou, Y.J. Cho, X. Liu, J.K. Furdyna, and M. Dobrowolska, unpublished.

²⁷ Amr M. Mahros, M.O. Luen, A. Emara, S.M. Bedair, E.A. Berkman, N.A. El-Masry, and J.M. Zavada, *Appl. Phys. Lett.* **90**, 252503 (2007).